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IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF

ARNO LANGE, ET AL.

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EXAMINER: TOOMER, CEPHIA D.

SERIAL NO: 10/089,064

:

FILED: APRIL 5, 2002

: GROUP ART UNIT: 1714

FOR: METHOD FOR PRODUCING
MANNICH ADDUCTS THAT CONTAIN
POLYISOBUTYLENE PHENOL

:

LETTER TO THE EXAMINER

c/o COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

Applicants submit herewith a copy of the Reply Brief filed by Applicants in the
Opposition proceedings of European Patent 1 226 188 B1. The European Patent corresponds
to the present application, U.S. Serial No. 10/089,064.

Respectfully submitted,

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Our ref.: M/40285-OPPO

Subject: **Opposition of Lubrizol Ltd. to EP 1 226 188 B1 (00971319.9-2109)
"Process for the preparation of polyisobutylene phenol-containing
Mannich adducts"**
Patent proprietor: BASF Aktiengesellschaft

In response to the communication according to Rule 57(1) EPC of 10.07.2005:

A. Requests

1. It is requested that the opposition to European Patent 1 226 188 B1 be rejected.
2. In the event that the Opposition Division does not grant this request during the written procedure, we request oral proceedings.

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B. Subject matter of the patent

The subject matter of claim 1 is a process for the preparation of polyisobutlenylphenol-containing Mannich adducts by

- a) alkylation of a phenol with polyisobutene having more than 70 mol% of vinylidene double bonds and a number average molecular weight of from 300 to 3000 at below about 50°C in the presence of an alkylation catalyst;
- b) reaction of the reaction product from a) with formaldehyde, an oligomer or a polymer of formaldehyde and at least one amine which has at least one secondary amino function and no primary amino function, or
- c) reaction of the reaction product from a) with at least one adduct of at least one amine which has at least one secondary or primary amino function and formaldehyde, an oligomer of formaldehyde or a polymer of formaldehyde or a formaldehyde equivalent.

The subclaims 2 to 9 relate directly or indirectly back to claim 1.

The subject matter of claim 10 is a Mannich adduct which is obtainable by a process as claimed in claim 1, first variant.

The subject matter of claim 11 is the use of a Mannich adduct as claimed in claim 10 as a detergent additive in fuel and lubricant compositions.

The subject matter of claim 12 is an additive concentrate comprising, in addition to conventional additive components, at least one Mannich adduct as claimed in claim 10 in amounts of from 0.1 to 99.9% by weight, preferably from 0.5 to 80% by weight.

The subject matter of claim 13 is a fuel composition which contains a main amount of liquid hydrocarbon fuel and an amount, having detergent activity, of at least one adduct as claimed in claim 10.

The subject matter of claim 14 is a lubricant composition containing a main amount of a liquid, semisolid or solid lubricant and an amount, having detergent activity, of at least one adduct as claimed in claim 10.

The subject matter of claim 15 is the use of a fuel composition as claimed in claim 13 as a gasoline or diesel fuel.

C. Comment on Article 100a) EPC

Re novelty:

Document D1 describes a composition which contains a Mannich adduct in combination with an alcohol and the use thereof as a detergent additive for hydrocarbon fuels (D1, Page 1, 1st paragraph). The Mannich adduct is obtained from (A) an alkylated hydroxyaromatic having a hydrocarbon radical which may have up to 400 carbon atoms, (B) an aldehyde or ketone and (C) a mixture of water and an amine which has at least one primary or secondary amino group (paragraph extending from bottom of page 1 to top of page 2 under "Summary of the Invention").

A process for the preparation of polyisobutylphenol-containing Mannich adducts, corresponding to variant 1 of claim 1 of the contested patent, in which, for alkylation, a phenol is reacted with a polyisobutene

- having a proportion of > 70 mol% of vinylidene double bonds,
- and a number average molecular weight of from 300 to 3000,
- at a temperature below about 50°C

and the reaction product thus obtained is reacted with at least one amine which has at least one secondary amino function and no primary amino function and with formaldehyde, an oligomer or a polymer thereof, and Mannich adducts obtainable by such processes, are not evident from the general disclosure of D1. Thus, very generally suitable hydrocarbon radicals of the hydroxyaromatics used for preparation of the Mannich adducts are listed on page 3, second paragraph to page 5, third paragraph of D1. The polyisobutene group which was picked out by the opponent and is derived from Ultravis®10 (D1, page 5, lines 1-3) is only one suitable embodiment of such a hydrocarbon radical among a multiplicity of others. In addition to formaldehyde, a multiplicity of further aldehydes and ketones are also mentioned as suitable aldehyde and ketone component (B) (page 5, last paragraph to page 6, 1st paragraph). The same also applies to the amine component (C) (D1, page 6, 2nd paragraph to page 8, last paragraph). Accordingly, it may be, *inter alia*, ammonia, primary or secondary amines, amines containing hydroxyl groups, polyamines, hydroxyalkyl-substituted alkylenamines and higher condensates. The special starting material combination of claim 1, variant 1, of the contested patent is not evident from the general disclosure of D1. The general disclosure of D1 also reveals nothing about the process conditions in the alkylation of the hydroxyaromatic. Moreover, the person skilled in the art could not have picked out individual aspects in isolation, such as the specific temperature range during the alkylation or the use of an alkylation catalyst, from the teaching of the examples and viewed them as general technical teaching for the alkylation reaction. The example of D1 (page 11) also does not anticipate the subject matter of variant 1 of claim 1 since ethylenediamine, i.e. an amine having two primary amino functions, is used therein.

Variant 2 of claim 1 of the contested patent, according to which a phenol alkylated with polyisobutene is reacted with at least one adduct of at least one amine which has at least one secondary or primary amino function and formaldehyde or an oligomer, polymer or equivalent thereof, is also not evident from the general disclosure of D1. Thus, D1 does not teach first

preparing an adduct from the amine component and the formaldehyde component and then reacting said adduct with a polyisobutarylphenol prepared as described above. Since the preparation of this polyisobutarylphenol (step a) of the process forming the subject of the contested patent is effected as in the case of variant 1, the above statements on the non-disclosed process conditions in the alkylation of the hydroxyaromatic apply here analogously. The working example of D1 also does not teach the reaction of polyisobutarylphenol with an adduct of amine and formaldehyde. Thus, in part B of the example, a mixture of polyisobutarylphenol and paraformaldehyde is initially taken and then a mixture of water and ethylenediamine is added and reacted to give the Mannich adduct.

The subject matter of claim 1 of the contested patent is therefore novel compared with D1. The same applies to the Mannich adducts based on an amine which has at least one secondary and no primary amino function as claimed in claim 10, the use of such a Mannich adduct as claimed in claim 11, an additive concentrate based on such a Mannich adduct as claimed in claim 12, pure compositions based on such a Mannich adduct as claimed in claim 13, lubricant compositions based on such a Mannich adduct as claimed in claim 14 and the use of a fuel composition based on such a Mannich adduct as claimed in claim 15.

Re Inventive step:

The present invention is concerned with the provision of a process for the preparation of polyisobutarylphenol-containing Mannich adducts for use as a detergent additive in fuel and lubricant compositions (contested patent, page 2, lines 1-5). D8, which likewise relates to reaction products of the Mannich reaction and the use thereof as a detergent in hydrocarbon fuels, is suitable as the closest prior art. Starting from D8, it is the object of the invention to provide a process for the preparation of polyisobutarylphenol-containing Mannich adducts having improved performance characteristics, in particular an

improved detergent effect. Surprisingly, it has now been found that this object is achieved by a process in which polyisobutene-substituted phenols obtained under special process conditions are reacted either with formaldehyde, oligo- or polymers of formaldehyde in the presence of a secondary amine or with an adduct of at least one amine with formaldehyde or another formaldehyde source, and the Mannich adducts obtainable by the first process variant.

The achievement of this object was also not obvious. Regarding the amine component used, D8 teaches the exclusive use of ethylenediamine. It is therefore precisely this feature that the person skilled in the art would not have varied. Consequently, D8 does not suggest to the person skilled in the art the first process variant of claim 1 of the contested patent, according to which an amine which has at least one secondary amino function but no primary amine function is used.

The second variant of the process forming the subject matter of the contested patent was also not obvious. Thus, the person skilled in the art had no reason for picking out the teaching of examples 1 and 2 of D1 in isolation and making it the starting point for the provision of improved Mannich adducts by subjecting the amine and the formaldehyde component to adduct formation. The teaching of the remaining examples would in any case not have been considered by the person skilled in the art since these are comparative examples. The second variant was therefore not obvious in view of a combination of D8 with D9. Thus, D9 (paragraph 8.6, page 261) does not distinguish in particular between the reaction of primary and secondary amines in the Mannich reaction. The reaction scheme a) at the bottom of page 261 is merely a text-book description of the reaction mechanism with the intermediates formed. Here, the person skilled in the art does not find the slightest suggestion and certainly not an explicit teaching that he should first form an adduct from amine component and formaldehyde component in the reaction of primary amines in the Mannich reaction. Furthermore, comparative example VB2 in table 2 on page 17 of the contested patent shows that, with fuel additives according to example 2 of D8, i.e. with Mannich adducts based

on ethylenediamine, satisfactory results in keeping valves clean are not obtained without prior adduct formation. The claimed subject matter is therefore based on an inventive step in view of a combination of D8 with D9.

The achievement of the object was also not obvious with regard to a combination of D3 and D4. D3 relates to fuel additives for gasoline fuels. This document is based on the object of reducing or of avoiding undesired valve deposits in electronically controlled direct-injection engines. (D3, page 1, lines 13-18). For achieving this object, D3 teaches the use of an additive mixture comprising a) a Mannich condensate and b) an oil-soluble poly(oxyalkylene) alcohol, glycol, polyol or a mono- or diether thereof. Neither the Mannich adduct as such nor the preparation of the alkyl-substituted hydroxyaromatic used for the Mannich reaction is considered to be particularly critical. Thus, regarding the alkyl group of the hydroxyaromatic, it is merely mentioned that it may be, for example, a polyalkyl radical which may be derived from 1-monoolefin polymers having a number average molecular weight of from 600 to 3000 (page 6, lines 1-4). The passage on page 11, line 4 which is used by the opponent as evidence of the use of a polyisobutetyl-substituted phenol relates to a comparative example (example 2) in which a polyisobutenamine (and not a Mannich adduct) is used as a fuel additive. Alkylation of the hydroxyaromatic with a polyisobutene, to say nothing of a highly reactive polyisobutene according to the definition of claim 1 of the contested patent (i.e. having a content of at least 70 mol% of vinylidene double bonds), is not disclosed. There are also no statements about the other process conditions of the alkylation reaction. In the case of the amine component (D3, page 6, lines 5-11) no distinction is made between amines having primary amino functions and amines having secondary amino functions. The document does not even contain a slightest hint about either using an amine which contains at least one secondary amino function and no primary amino function in the preparation of the Mannich adducts or first forming an adduct from amine component and formaldehyde component.

The combination of D3 with D4 used by the opponent does not lead to the claimed achievement simply because D4 relates to a process for the preparation of a polyisobutetyl-substituted hydroxyaromatic compound and not to a process for the preparation of a Mannich adduct. The polyisobutyl-substituted hydroxyaromatics disclosed in D4 serve in turn for use as fuel additives (e.g. D4, column 4, lines 65-68). Starting from D3, the person skilled in the art had no reason to use the teaching of D4 for achieving the object set, since he cannot assume, at least not with a safe expectation of success, that the advantageous properties which the alkylated hydroxyaromatics of D4 have when used as fuel additives would occur again in the respective end product after a fundamental chemical modification by an amino alkylation according to Mannich. To this extent, the facts of the case also differ fundamentally from those of T 94/84 which is used by the opponent and in which it was very clear that the product of the document used for the combination is one which is very evidently suitable for achieving the object set and where this alternative would have been obvious to the person skilled in the art. There can therefore be no question of this in the case of a required fundamental structural modification as arises from a combination of the teaching of D3 with that of D4. Even in the case of a combination of the teachings of D3 and D4, the subject matter of claim 1 of the contested patent is nevertheless not arrived at since the special reaction procedure as a function of the amine component used, corresponding to reaction steps b) and c), is not evident from any of the documents.

The additional combination of D3 and D4 with D5 also does not suggest the claimed subject matter. It is firstly questionable as to whether D5 is at all prior art to be taken into account for the question of inventive step. D5 itself was introduced on 07.18.2001, i.e. after the date of filing of the contested patent, into the opposition appeal proceedings against EP 628 022. This document is a "Pilot Plant Report" from the year 1956, in which there is no evidence that it was ever made available to the public before 07.18.2001. However, even if D5 was available to the public, it would never have revealed to the person skilled in the art the teaching that he should use, for the preparation of polyisobutetyl-containing Mannich adducts, a polyisobutetylphenol which was

obtained by alkylation of a phenol with highly reactive polyisobutene at a temperature of less than 50°C. Thus, the alkylation used by the opponent on page 2 of D5 describes the reaction of phenol with an unspecified polybutene at a temperature of from 120 to 130°F (48.92-54.48°C). To derive from this that the alkylation of phenol with polyisobutene at temperatures of less than 50°C is part of the standard knowledge of the average person skilled in the art is odd. Furthermore, it is also not true that polyisobutenes, independently of their content of vinylene double bonds, can be used at temperatures of less than 50°C for the alkylation of phenols. Thus, the Friedel-Crafts alkylation of hydroxyaromatics having internal double bonds is successful only under drastic conditions and leads to an undesired depolymerization as a secondary reaction.

The achievement of the object was also not obvious in view of a combination of D3 with D6 or D7. As already mentioned above, D3 does not teach the use of polyisobutenylphenols, as are obtainable by alkylation of a phenol with highly reactive polyisobutene under the special process conditions of stage a) of claim 1 of the contested patent, for the preparation of Mannich adducts. D3 furthermore does not teach subsequently reacting these polyisobutenylphenols with an amine which has at least one secondary amino function and no primary amino function or first forming an adduct from the amine component and formaldehyde component and reacting said adduct with the polyisobutenylphenol. This is also not suggested to the person skilled in the art by documents D6 and D7. Thus, these documents are different press texts which merely reveal that a chlorine-free polybutene having a higher proportion of reactive double bonds than was known up to the date of publication of the press texts is sold by BP Chemicals under the name Ultravis. These documents provide no specific technical teaching on the use of these polyisobutenes in the alkylation of phenols nor about the subsequent reaction thereof to give Mannich adducts. A combination of D3 with one of the press texts reproducing the documents D6 and D7 thus does not lead to the subject matter of the claim.

The claimed subject matter is also based on an inventive step. The request to refuse the opposition and to maintain the patent in the extent granted is therefore justified.

(Jens Wortmann)

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